

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants : Juliana G. Serafin, et al. Confirmation No. 7169
Appln. No. : 10/573,694 Examiner: Zimmer
Filed : March 27, 2006 Group Art Unit: 1793
Docket No. : 62397A
Title : CATALYSTS HAVING ENHANCED STABILITY, EFFICIENCY
AND/OR ACTIVITY FOR ETHYLENE OXIDE PRODUCTION

Declaration Under 37 C.F. R. § 1.132

The undersigned, Dr. Juliana G. Serafin, citizen of the United States of America and residing in Charleston, West Virginia, declare and say that:

1. I am a 1982 graduate of the West Virginia University, Morgantown, WV with a Bachelor of Science degree in Chemistry; a 1984 graduate of Harvard University with a Master of Arts in Chemistry; and a 1989 graduate of Harvard University with a Ph.D. in Physical Chemistry.

2. From October 1989 to August 2008, I was employed by Union Carbide Company ("UCC") in South Charleston in research and development of ethylene oxide catalysts. The positions I held included: Senior Chemist, Project Scientist, Research Scientist, and Research Leader. My responsibilities included the discovery and implementation of new ethylene oxide catalyst formulations as well as modification of catalyst carriers.

3. In the fall of 2008, I began teaching at University of Charleston as an Assistant Professor of Chemistry.

4. I am currently an inventor on three US published applications in the field of ethylene oxide catalysis.

5. I have read the above referenced patent application, the Office Action dated May 2, 2008 and the prior art reference, Buffum, U.S. Patent No. 5,145,824 cited by the Examiner.

6. During my employment with UCC, I requested that a sample be made of alpha alumina carrier containing 1% granular zirconium silicate as a starting material

(Carrier A) and an alpha alumina carrier containing 1% granular zirconia (baddeleyite, a monoclinic form) as a starting material (Carrier B). Upon information and belief, the samples were prepared using the procedures described for the preparation of carriers using pseudoboehmite on pages 19 and 20 of the present application, including calcination at a temperature of between 1000 and 1400°C. X-ray Diffraction analysis of Carrier B showed a monoclinic form of zirconia was present.

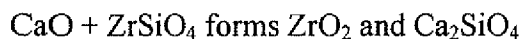
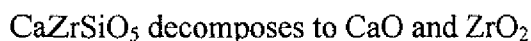
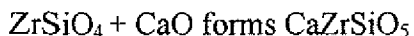
7. Carrier A and Carrier B were used to prepare Catalysts A and B, respectively, using the procedures described on pages 20 and 21 of the present application. According to X-Ray Fluorescence testing, the amounts of silver and promoters on Catalyst A were 32.8 % Ag by weight, 550 ppm Cs, 154 ppm SO₄, and 99 ppm Mn. According to X-Ray Fluorescence testing, the amounts of silver and promoters on Catalyst B were 34% by weight, 581 ppm Cs, 138 ppm So₄ and 108 ppm Mn.

8. Catalysts A and B were tested under the following oxygen process conditions in autoclaves: inlet conditions of 30.0% ethylene, 6.5% CO₂, 8.0% O₂, 0.50% ethane (all mole %) and 3.5 ppm ECl. The reactor pressure was maintained at 275 psig. The inlet flow rate was 22.6 SCFH nitrogen at STP, and catalyst charge size was 63.5 g.

9. As shown by the data below, the initial performance of Catalyst B was less active and less selective than Catalyst A.

Catalyst	Day	Eff.% @ 2.0% EO	Temp.°C @ 2.0% EO
A	12	79.5	237
B	9	79.1	247
	15	78.7	247

10. As explained in Blumenthal, the decomposition of zirconium silicate in the presence of calcium is illustrated in the following equations:

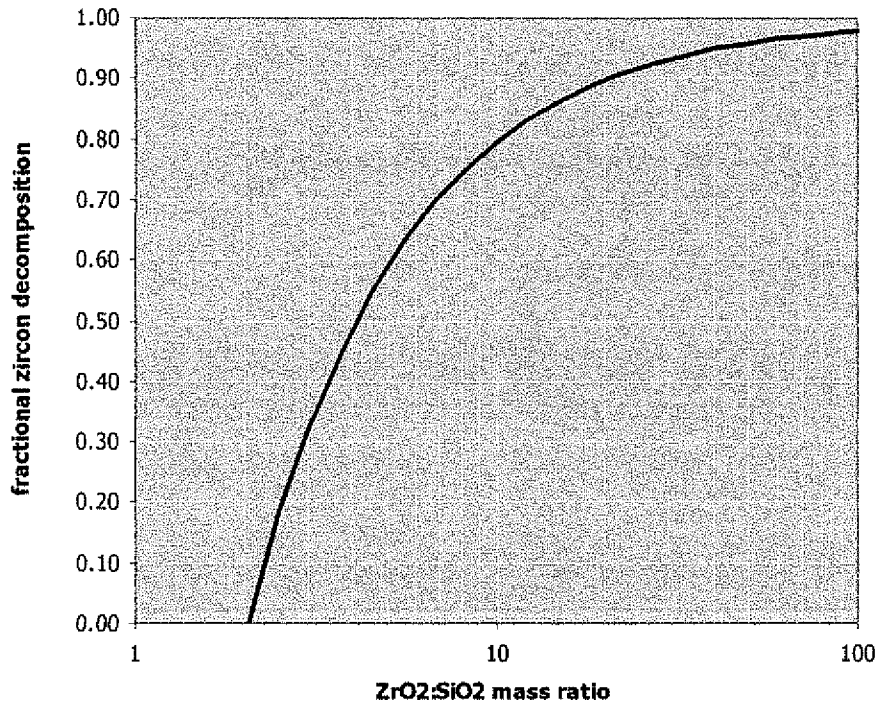


11. The experimental work on the zirconium silicate modified carrier further included the examination of the potential effects of the presence of calcium on the decomposition

of zirconium silicate during calcination of the carrier precursor. Upon information and belief, the carrier samples C and D below were prepared using the procedures described for the preparation of carriers using pseudoboehmite on pages 19 and 20 of the present application, including calcination at a temperature of between 1000 and 1400°C, in particular at a temperature of between 1000 and 1250°C. In each instance, 1000 ppm of calcium was mixed with the precursor. We learned that when the 1000 ppm Ca was added to the mix, the Si was almost completely stripped from the final product.

12. The table below contains data corresponding to zircon-modified Carriers C and D also doped with 1000 ppm Ca, and comparison Carrier E modified with zircon but no added Ca. The plot below allows conversion of analyzed $\text{ZrO}_2\text{:SiO}_2$ weight ratios to the fractional zircon decomposition estimates, assuming that all of the Si liberated by decomposition of the original zirconium silicate compound during firing is volatilized and thus “missing” from the finished carrier. That is, a ratio of 2.05 would correspond to zircon itself, so any higher ratios of ZrO_2 to SiO_2 would reflect depletion of Si. In contrast to Carrier E (no added Ca), the samples with the presence of 1000 ppm Ca show nearly a 100% loss of zircon. Furthermore, when samples similar in composition to Carrier E are analyzed by X-ray Diffraction, alpha-alumina and zircon phases are detected, and when a zirconium oxide phase is also seen to be present, only the monoclinic form of zirconia is observed.

Carrier	Zircon (wt %)	Calcium (ppm)	wt% ZrO_2	wt% SiO_2	$\text{ZrO}_2\text{: SiO}_2$	Fractional zircon decomposition
C	2%	1000	1.65	0.03	55.0	0.96
D	2%	1000	1.46	0.02	73.0	0.97
E (control)	2%	—	1.62	0.76	2.1	0.03



12. I calculated the Zr:Si ratios for the carrier in the presence of Ca and found them to be more than an order of magnitude higher than those in comparable carriers without the added Ca. X-Ray Diffraction analysis revealed increased levels of zirconia in the fired carrier containing the added calcium as compared to the carrier without the added Ca.

13. I declare further that all statements made herein of my knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Respectfully submitted,

Juliana G. Serafin
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April 27, 2009